

Practitioner's Docket No.: 791\_169

PATENT

IN THE UNITED STATES DESIGNATED OFFICE (DO/US)

PCT/JP01/01562	01 March 2001	02 March 2000
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED

TITLE OF INVENTION

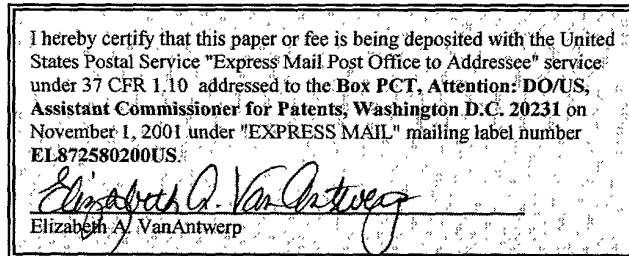
ZEOLITE SHAPED BODY, ZEOLITE LAYERED INTERMEDIATE BODY, ZEOLITE LAYERED COMPOSITE, AND PRODUCTION METHOD FOR THE SAME

APPLICANT(S) FOR DO/US

Toshihiro TOMITA and Shinji NAKAMURA

Box PCT  
Assistant Commissioner for Patents  
Washington, D.C. 20231

Attention: DO/US



PRELIMINARY AMENDMENT

Sir:

Prior to examination, Applicants wish to amend the subject application as follows:

In the Claims:

Please cancel claims 1-24 without prejudice or disclaimer in favor of new claims 25-48.

25. (New) A porous zeolite shaped body of a zeolite, characterized in that said porous zeolite shaped body is made of a completely crystallized zeolite composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole.

26. (New) A porous zeolite shaped body of a zeolite, characterized in that the porous zeolite shaped body is made of a zeolite still under crystallization and composed of

tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.02 to 0.12 by mole.

27. (New) A zeolite intermediate body, characterized in that the zeolite shaped body as claimed in Claim 25 contains further a template, and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is formed thereon.

28. (New) A zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon, characterized in that the composite is produced by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as claimed in Claim 27.

29. (New) A method for producing a zeolite layered composite, characterized by layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite shaped body of a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole and containing a template therein on said zeolite shaped body, and simultaneously removing the template from said zeolite membrane and said zeolite shaped body by calcining the resulting layered product to obtain a zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon.

30. (New) A method for producing a zeolite layered composite, characterized by layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite shaped body of a zeolite still under crystallization and composed of

tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.02 to 0.12 by mole and containing a template therein on said zeolite shaped body, and simultaneously removing a template from said zeolite membrane and said zeolite shaped body by calcining the resulting layered product to obtain a zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon.

31. (New) A porous zeolite shaped body of a zeolite, characterized in that the porous zeolite shaped body has an average particle diameter of 1.0 mm or larger, a bending strength of 1.5 MPa or higher, and a difference in pressure between a feed side and a permeation side of 1.0 atmospheric pressure or lower at 10 ml/cm<sup>2</sup>·min of helium gas permeation flux when a thickness of the porous zeolite shaped body is adjusted to be 1.8 mm.

32. (New) A zeolite layered intermediate body, characterized in that the zeolite shaped body as claimed in Claim 31 contains, further a template and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is layered thereon.

33. (New) A zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon, characterized in that the zeolite layered composite is formed by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as claimed in Claim 32.

34. (New) A method for producing a zeolite shaped body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that mixing ratios  $[\text{TPAOH}/(\text{TPAOH} + \text{TPABr})]$  and  $\text{TPABr}/(\text{TPAOH} + \text{TPABr})$  of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% by mole and 1 to 100% by mole, respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, and subjecting thus shaped body to crystallization treatment.

35. (New) A method for producing a zeolite shaped body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, and subjecting thus shaped body to crystallization treatment.

36. (New) A method for producing a zeolite layered intermediate body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that mixing ratios  $[\text{TPAOH}/(\text{TPAOH} + \text{TPABr})]$  and  $\text{TPABr}/(\text{TPAOH} + \text{TPABr})$  of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% by mole and 1 to 100%, respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to said prepared solution, and forming a template-containing zeolite membrane on said zeolite

shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.

37. (New) A method for producing a zeolite layered intermediate body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting the shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.

38. (New) A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that the mole ratio of mixing ratios  $[\text{TPAOH}/(\text{TPAOH} + \text{TPABr})]$  and  $[\text{TPABr}/(\text{TPAOH} + \text{TPABr})]$  of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% and 1 to 100%, respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said

zeolite shaped body and said template-containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

39. (New) A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

40. (New) A porous zeolite shaped body of a zeolite, characterized in that area of parts (sound parts) where respective particles are clearly observed by grain boundary fracture among particles composing the zeolite shaped body in microstructure observation of the fractured surface of the shaped body occupies 70% or more in the entire area of the fractured surface.

41. (New) A zeolite layered intermediate body, characterized in that the zeolite shaped body as claimed in Claim 40 further contains a template, and a template-containing zeolite membrane having a composition same as or similar to that of the shaped body is formed on the shaped body.

42. (New) A zeolite layered composite comprising a zeolite shaped body and a zeolite membrane formed thereon, characterized in that the zeolite layered composite is produced by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as claimed in Claim 41.

43. (New) A method for producing a zeolite shaped body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by kneading the solution, wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus obtained dried granular substance, and subjecting thus obtained substance to crystallization treatment to obtain a zeolite shaped body.

44. (New) A method for producing a zeolite shaped body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, and subjecting thus obtained gel to crystallization treatment to obtain a zeolite shaped body.

45. (New) A method for producing a zeolite layered intermediate body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by kneading the solution,

wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane.

46. (New) A method for producing a zeolite layered intermediate body characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 by mole to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting to thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite membrane.

47. (New) A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by kneading the solution, wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus



obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite membrane, and then simultaneously removing a template by calcining the layered body.

48. (New) A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite membrane, and then simultaneously removing a template by calcining the layered body.

**In the Abstract:**

Please rewrite the abstract as follows;

**ABSTRACT**

A porous zeolite shaped body made of a completely crystallized zeolite or a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole, having an average particle diameter of 1.0 mm or larger, a bending strength of 1.5 MPa or higher, and a difference in pressure between a feed side and a permeation side of 1.0 atmospheric pressure or lower at 10 ml/cm<sup>2</sup>·min of helium gas permeation flux when a thickness of the porous zeolite shaped body is adjusted to be 1.8 mm, and having 70% or more of the area of the parts where respective particles are clearly observed by grain boundary fracture among particles composing the shaped body in the entire area of the fractured surface in microstructure observation of the fractured surface of the shaped body itself.

**REMARKS**

Claims 25-48 are pending herein. Claims 1-24 have been canceled without prejudice or disclaimer in favor of new claims 25-48. No new matter has been added. Applicants believe the case is now in condition for examination.

Attached hereto as page 12 titled VERSION WITH MARKINGS TO SHOW CHANGES MADE is a marked-up version of the Abstract showing changes made thereto.

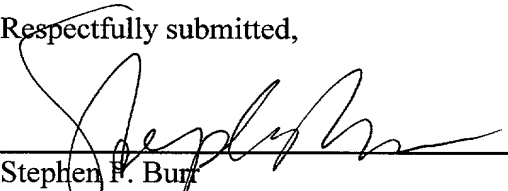
If the Examiner believes that contact with applicants' attorney would be advantageous toward the disposition of this case, he is herein requested to call applicants' attorney at the phone number noted below.

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,

November 1, 2001

Date

  
Stephen F. Burr  
Reg. No. 32,970

SPB/eav

BURR & BROWN  
P.O. Box 7068  
Syracuse, NY 13261-7068

Customer No.: 025191  
Telephone: (315) 233-8300  
Facsimile: (315) 233-8320

**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

**In the Abstract:**

The abstract has been amended as follows:

**ABSTRACT**

A porous zeolite shaped body ~~of a zeolite is characterized in that the porous zeolite shaped body is made of a completely crystallized zeolite or a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole; a zeolite shaped body has~~ having an average particle diameter of 1.0 mm or larger, a bending strength of 1.5 MPa or higher, and a difference in pressure between a feed side and a permeation side of 1.0 atmospheric pressure or lower at 10 ml/cm<sup>2</sup>·min of helium gas permeation flux when a thickness of the porous zeolite shaped body is adjusted to be 1.8 mm<sup>2</sup>, and ~~a zeolite shaped body has~~ having 70% or more of the area of the parts ~~(the sound parts)~~ where respective particles are clearly observed by grain boundary fracture among particles composing the shaped body in the entire area of the fractured surface in microstructure observation of the fractured surface of the shaped body itself.